

PATENT ABSTRACTS OF JAPAN

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(54) CURABLE COMPOSITION

(57)Abstract:

PROBLEM TO BE SOLVED: To obtain the subject composition capable of being cured by a hydrosilylation reaction and excellent in adhesivity by including a specific polymer, a specific curing agent, a specific catalyst: a silane coupling agent, and two kinds of specific compounds.

SOLUTION: This curable composition comprises (A) an isobutylene polymer containing at least one alkenyl group capable of being hydrosilylated in the molecule, (B) a curing agent containing at least two hydrosilyl groups in the molecule (e.g. an organo hydrogen polysiloxane), (C) a hydrosilylation catalyst, (D) a silane coupling agent preferably containing an epoxy group and a (meth) acryl group in the molecule, (E) an organic aluminum compound and/or an organic titanium compound, (F) a polyvalent alkoxysilane (condensate) (e.g. a tetraalkoxy silane), and, if necessary, (G) a plasticizer, an inorganic filler, a storage stability-improving agent, etc.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] this invention relates to the hardenability constituent containing the saturated-hydrocarbon system polymer hardened by the hydrosilylation reaction.

[0002]

[Description of the Prior Art] Various kinds of things are developed as a hardenability liquefied constituent which generates the rubberlike substance by hardening conventionally. As the hardening system which was excellent in deep part hardenability especially. Into the end or the chain, what constructs a bridge by the ORGANO hydrogen polysiloxane which has the hydrogen atom which combines with a silicon atom the organopolysiloxane which has an average of two pieces or a vinyl group beyond it in one molecule in [an average of two or more] 1 molecule is developed, and it is used as a silicone system sealing material or potting material using the outstanding weatherability, thermal resistance, and water resistance. However, this hardening system has high cost and the use is restricted from the reason of being easy to generate mold with a bad adhesive property. The hardenability constituent which constructs a bridge into a molecule as a system which does not use such expensive organopolysiloxane on the other hand with the curing agent which has the hydrogen atom which combines with two silicon atoms in a molecule the saturated-hydrocarbon system polymer which has at least one alkenyl machine is indicated by JP,2-75644,A, JP,3-181565,A, etc. The hardened material of this constituent has the outstanding weatherability, thermal resistance, low moisture permeability, etc., and use for a broad use is expected. However, although the hardenability constituent whose polymer is a saturated-hydrocarbon system polymer is excellent in hardenability or the mechanical characteristic as rubber, its adhesive property to a metal, plastic material, etc. is inadequate. Moreover, although JP,4-185687,A was shown the adhesives constituent which added the silane coupling agent to the above-mentioned constituent for the purpose of these adhesive improvement, the adhesive improvement effect was [of a certain thing] inadequate. In addition to the silane coupling agent, as a constituent in which the outstanding adhesive property is shown, Japanese Patent Application No. 8-134729 is shown the adhesives constituent which added the organoaluminium compound and/or the organic titanium compound.

[0003]

[Problem(s) to be Solved by the Invention] The purpose of this invention is in the hardenability constituent which can be hardened according to addition mold curing using the saturated-hydrocarbon system polymer excellent in low moisture permeability, low hygroscopicity, low permeability, thermal resistance, weatherability, electric insulation, and oscillating absorptivity to improve an adhesive property further.

[0004]

[Means for Solving the Problem] When this invention persons applied the saturated-hydrocarbon system polymer which makes an isobutylene representation to an addition mold-curing system, they found out that the hardened material which consists of a saturated-hydrocarbon system excellent in the adhesive property was obtained by adding a silane coupling agent, an organoaluminium compound and/or an organic titanium compound and multiple-valued alkoxysilane, and/or its condensation product.

[0005] That is, this invention relates to the hardenability constituent containing the (curing agent C) hydrosilylation catalyst (D) silane-coupling-agent (E) organoaluminium compound which contains at least two hydrosilyl machines in the isobutylene system (polymer B) molecule which contains the alkenyl machine in which at least one hydrosilylation reaction is possible in the (A) molecule, organic titanium compound (F) multiple-valued alkoxysilane, and/or its condensation product.

[0006]

[Embodiments of the Invention] The (A) component used for this invention is a saturated-hydrocarbon system polymer which has in a molecule the alkenyl machine in which at least one hydrosilylation reaction is possible. Here, a saturated-hydrocarbon system polymer is a concept meaning the polymer which does not contain carbon-carbon unsaturated bonds other than a ring substantially, and means that the repeat unit which constitutes the principal chain except this alkenyl machine consists of saturated hydrocarbons. Moreover, it will not be restricted especially if the alkenyl machine in which a hydrosilylation reaction is possible is a basis including carbon-carbon double combination which has activity to a hydrosilylation reaction. As an alkenyl machine, ring type unsaturation hydrocarbon groups, such as aliphatic-unsaturated-hydrocarbon machines, such as a vinyl group, an allyl group, a methyl vinyl group, a propenyl machine, a butenyl group, a pentenyl machine, and a hexenyl

machine, a cyclo propenyl machine, a cyclo butenyl group, a cyclo pentenyl machine, and a cyclohexenyl group, are mentioned. As for the (A) component, in this invention, it is desirable to have 1-10 alkenyl machines in 1 molecule. Furthermore, as for the (A) component in this invention, it is desirable to introduce into the polymer end the alkenyl machine in which the above-mentioned hydrosilylation reaction is possible. Thus, when an alkenyl machine is in a polymer end, it is desirable from points, like the rubber-like hardened material of the high elongation in high intensity becomes effective mesh **** of the hardened material finally formed increases, and is easy to be obtained. (A) The polymer which makes the skeleton of the saturated-hydrocarbon system polymer which is a component (1) It carries out a polymerization, using the olefinic compound of the carbon numbers 1-6, such as ethylene, a propylene, 1-butene, and an isobutylene, as the main monomer. (2) Although it can obtain by the method of hydrogenating after making diene series, such as a butadiene and an isoprene, homopolymerize or carrying out copolymerization of the above-mentioned olefinic compound and the diene series It is desirable that it is the point of being able to make [many] the number of end functional groups to the isobutylene system polymer, hydrogenation poly-butadiene system polymer, or hydrogenation polyisoprene system polymer which is easy to introduce a functional group into an end and which is easy to carry out molecular weight control.

[0007] the monomeric unit with which all the monomeric units may be formed from the isobutylene unit, and the aforementioned isobutylene system polymer has an isobutylene and copolymerization nature -- an isobutylene system polymerization -- the inside of the body is desirable and you may contain in 20% or less of range preferably especially 30% or less still more preferably below 50% (% of the weight and the following -- the same) As such a monomer component, the olefin of carbon numbers 4-12, vinyl ether, an aromatic vinyl compound, vinyl silanes, and arylsilane are mentioned, for example. As an example of such a copolymer component, for example 1-butene, 2-butene, A 2-methyl-1-butene, a 3-methyl-1-butene, a pentene, 4-methyl-1-pentene, A hexene, a vinyl cyclohexane, the methyl vinyl ether, ethyl vinyl ether, An isobutyl vinyl ether, styrene, an alpha methyl styrene, dimethyl styrene, p-t-butoxy styrene, p-hexenyloxy styrene, p-aryloxy styrene, p-hydroxy styrene, beta-pinene, an indene, a vinyl dimethyl methoxy silane, A vinyl trimethyl silane, divinyl dimethoxysilane, divinyl dimethylsilane, 1, 3-divinyl - 1, 1, 3, and 3-tetramethyl disiloxane, a TORIBI nil methylsilane, A tetravinyl silane, an allyl-compound dimethyl methoxy silane, an allyl-compound trimethyl silane, Diaryl dimethoxysilane, diaryl dimethylsilane, gamma-methacryloyl oxypropyl trimethoxysilane, gamma-methacryloyl oxypropyl methyl dimethoxysilane, etc. are mentioned.

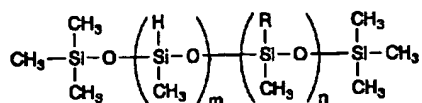
[0008] You may make other monomeric units contain besides the monomeric unit used as a principal component like the case of the above-mentioned isobutylene system polymer in the aforementioned hydrogenation poly-butadiene system polymer or other saturated-hydrocarbon system polymers. moreover, a monomeric unit with which the double combination after a polymerization like a butadiene and a polyene compound like an isoprene remains in the saturated-hydrocarbon system polymer used as a (A) component in this invention in the range in which the purpose of this invention is attained -- small quantity -- you may make it contain in 10% or less of range preferably

[0009] As for the number average molecular weight (the GPC method, polystyrene conversion) of an isobutylene system polymer, a hydrogenation polyisoprene, or a hydrogenation poly-butadiene system polymer, it is desirable the aforementioned saturated-hydrocarbon system polymer and that it is 100000 or less and further 500 to about 100000, and it is preferably desirable from the point of the ease of dealing with it etc. that it is what has especially the liquefied object which is 1000 to about 40000, and a fluidity.

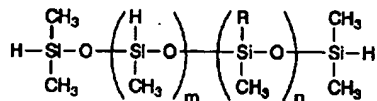
[0010] Moreover, the method of making the compound which has an unsaturation machine react to the polymer which has functional groups, such as a hydroxyl group which is indicated by JP,3-152164,A and JP,7-304909,A each official report, as the manufacture method of the (A) component of this invention, and introducing an unsaturation machine into a polymer is raised. Moreover, the method of using together the further aforementioned ARUKENIRU machine introduction method after performing the method of performing a Friedel Crafts reaction, a bottom allyl-compound of Lewis-acid existence trimethyl silane, etc. the method and the various phenols which perform a substitution reaction, and the Friedel Crafts reaction to introduce an unsaturation machine into the polymer which has a halogen atom and introducing a hydroxyl group etc. is raised. [an ARUKE nil phenyl ether,] The method of introducing an unsaturation machine at the time of the polymerization of a monomer which is furthermore indicated by U.S. Pat. No. 4316973, JP,63-105005,A, and JP,4-288309,A is also possible.

[0011] As a curing agent which is the (B) component of this invention, if at least two hydrosilyl machines are contained in a molecule, there will be no limit. Here, one SiH basis is put in one hydrosilyl basis. Therefore, when two hydrogen atoms have combined with the same Si, it calculates with two hydrosilyl bases. (B) As a component, although the ORGANO hydrogen polysiloxane is desirable, it is mentioned to one. If the ORGANO hydrogen polysiloxane said here points out the polysiloxane which has a hydrocarbon group or a hydrogen atom on Si atom and the structure is shown concretely [0012]

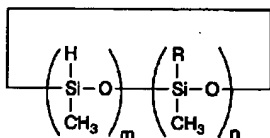
[Formula 1]



(mは2以上の整数、nは0以上の整数で、mとnの和は4以上50以下、Rは炭素数2～20のメチル基、フェニル基等の炭化水素基)



(mは2以上の整数、nは0以上の整数で、mとnの和は4以上50以下、Rは炭素数2～20のメチル基、フェニル基等の炭化水素基)

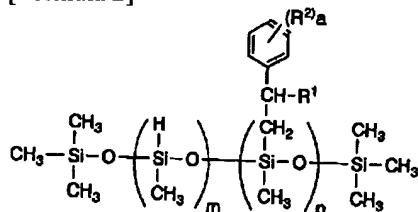


(mは2以上20以下の整数、nは0以上18以下の整数で、mとnの和は2以上20以下、Rは炭素数2～20のメチル基、フェニル基等の炭化水素基)

[0013] The shape of a chain shown by **** and an annular thing are mentioned. The following are especially desirable if a point with little possibility of spoiling the compatibility to various organic polymers, such as the (A) component of the hydrosilyl machine content curing agent which is the (B) component of this invention among various kinds of above-mentioned hydrosilyl machine content machines, is taken into consideration.

[0014]

[Formula 2]



(mは2以上の整数、nは0以上の整数で、mとnの和は4以上50以下、R¹は水素原子又はメチル基を表し、R²は炭素数1～4のアルキル基を表し、複数のR²が存在するときそれらは同一であっても異なってもよい。aは0～5の整数である。)

[0015] Moreover, although what is necessary is just to be in [two] 1 molecule at least about the number of the hydrosilyl machine contained in these (B) components, 2-40 pieces are desirable. If there is less number of this hydrosilyl machine than 2 when stiffening the constituent of this invention by the hydrosilylation reaction, hardening will cause poor hardening in many cases late. Moreover, if the number of this hydrosilyl machine increases more than 40, the stability of the curing agent which is the (B) component becomes bad, a lot of hydrosilyl machines will remain in a hardened material, and after the upper hardening will cause a void and a crack.

[0016] About the hydrosilylation catalyst which is the (C) component of this invention, there is especially no limit and it can use arbitrary things. thing; platinum-vinyl siloxane complex {which made support, such as a chloroplatinic acid, a simple substance of platinum, an alumina, a silica, and carbon black, support solid-state platinum when illustrating concretely -- for example Pt_n(ViMe₂SiOSiMe₂Vi)_n and Pt[(MeViSiO)₄]_m}; platinum-phosphine complex {-- for example Pt(PPh₃)₄ and Pt(PBu₃)₄}; platinum-phosphite complex {-- for example Pt[P(OPh)₃]₄ and Pt[P(OBu)₃]₄ (the inside of a formula, and Me -- a methyl group --) In Bu, a butyl and Vi express a vinyl group, Ph expresses a phenyl group, and n and m express an integer. The platinum alcoholate catalyst indicated in the platinum-hydrocarbon complex indicated in Pt(acac)₂, Ashby's and others U.S. patent 3159601st, and the No. 3159662 specification and Lamoreaux's and others U.S. Pat. No. 3220972 specification is also mentioned.

[0017] Moreover, as an example of catalysts other than a platinum compound, RhCl(PPh₃)₃, RhCl₃, Rh/aluminum 2O₃, RuCl₃, IrCl₃, FeCl₃ and AlCl₃, PdCl₂·2H₂O, NiCl₂, TiCl₄, etc. are mentioned. These catalysts may be used independently,

and even if it uses two or more sorts together, they are not cared about. A chloroplatinic acid, a platinum-olefin complex, a platinum-vinyl siloxane complex, and Pt(acac)₂ grade are desirable from the point of catalytic activity. Although there is especially no limit as an amount of catalysts, it is good to use in the range of 10-1 to ten to 8 mol to one mol of ARUKENIRU bases in the (A) component. It is good to use in the range of 10-2 to ten to 6 mol preferably. Moreover, it is better not to use ten - one mols or more, since a hydrosilylation catalyst is generally expensive, and it is corrosive, and hydrogen gas is generated in large quantities and a hardened material may foam.

[0018] In this invention, since a hardenability constituent hardens by the addition reaction of an Si-H basis to the ARUKENIRU machine using the noble metal catalyst, a cure rate is very quick, and when performing line production, it is convenient. The silane coupling agent which has at least one functional group chosen from an epoxy group, an methacrylic machine, an acrylic machine, an isocyanate machine, an isocyanurate machine, a vinyl group, and a carbamate machine into a molecule and silicon atomic-union alkoxy group as a silane coupling agent which is the (D) component of this invention is desirable. About the aforementioned functional group, it is desirable that an epoxy group, an methacrylic machine, and an acrylic machine are especially in a molecule from hardenability and an adhesive point especially. If it illustrates concretely, as an organic silicon compound which has an epoxy functional group and a silicon atomic-union alkoxy group, 3-glycidoxypropyltrimethoxysilane, 3-glycidoxypropyltriethoxysilane, 2-(3, 4-epoxycyclohexyl) ethyl trimethoxysilane, and 2-(3, 4-epoxycyclohexyl) ethyltriethoxysilane will be mentioned. Moreover, as an organic silicon compound which has an methacrylic machine or an acrylic machine, and a silicon atomic-union alkoxy group, 3-methacryloxy propyl trimethoxysilane, 3-methacryloxypropyl triethoxysilane, 3-acryloxypropyltrimethoxysilane, 3-acryloxypropyltriethoxysilane, meta-chestnut ROKISHI methyl trimethoxysilane, meta-KURIROKISHI methyl triethoxysilane, acryloxy methyl triethoxysilane, and acryloxy methyl triethoxysilane are mentioned.

[0019] Moreover, the chelate organic compound made into the useful catalyst of the hydrolysis condensation reaction of an alkoxy silyl machine as an organoaluminum compound which is the (E) component of this invention is desirable. Aluminum acetylacetonate etc. is mentioned as an example. Moreover, the organic titanium compound which has Ti-O-C combination made into the useful catalyst of the hydrolysis condensation reaction of an alkoxy silyl machine as an organic titanium compound which is the (E) component of this invention is desirable. As an example, general titanate coupling agents, such as what has residues, such as an oxy-acetic acid besides tetrapod alkoxy titanium, such as tetraisopropoxy titanium and tetrabutoxytitanium, and ethylene glycol, can be used.

[0020] Moreover, various alkoxy silane can be used as multiple-valued alkoxy silane which is the (F) component of this invention. These alkoxy silane is alkoxy silane which does not have functional groups, such as an epoxy group, an methacrylic machine, an acrylic machine, an isocyanate machine, an isocyanurate machine, a vinyl group, and a carbamate machine, in a molecule. Specifically, trialkoxy silane, such as tetrapod alkoxy silane, such as a tetramethoxy silane, a tetrapod ethoxy silane, a tetrapod (n-propyl) silane, and a tetrapod (n-butyl) silane, and a condensation product of those or methyl triethoxysilane, and ethyltriethoxysilane, and the condensation product of those can be used. In these, the trialkoxy silane which saturation alkyl groups, such as tetrapod alkoxy silane and a methyl group, replaced, or these condensates are desirable.

[0021] Moreover, to the hardenability constituent of this invention, various plasticizers and various inorganic fillers can be further added according to the purpose. Although the plasticizer which adds as a plasticizer in order to improve the fluidity of this constituent, and is generally used can be used, the good thing of the saturated-hydrocarbon system polymer used for this invention and compatibility is desirable. As an example of a plasticizer, although a polybutene, a hydrogenation polybutene, alpha-methyl-styrene oligomer, liquid polybutadiene, hydrogenation liquid polybutadiene, a paraffin oil, a naphthene oil, atactic polypropylene, etc. are mentioned, for example, hydrocarbon system compounds, such as a hydrogenation polybutene which does not include a unsaturated bond preferably in it, hydrogenation liquid polybutadiene, a paraffin oil, a naphthene oil, and atactic polypropylene, are desirable.

[0022] Moreover, as an inorganic filler, various kinds of inorganic fillers, such as a calcium carbonate, talc, a silica, and carbon black, can be used. However, in the use, if many moisture is contained in a constituent, for example, the hardenability constituent of this invention must take into consideration the existence of the prevention to a hydrosilylation reaction -- there is possibility that side reaction etc. will occur in hardening reaction time -- in order to use hardening by the hydrosilylation reaction.

[0023] A preservation stability improvement agent can be used in order to improve the preservation stability of the hardenability constituent of this invention furthermore. It is not limited especially that to be the usual stabilizer known as a preservation stabilizer of the (B) component of this invention as this preservation stability improvement agent, and what is necessary is just what attains the desired end. Specifically, the compound containing an aliphatic unsaturated bond, an organic phosphorous compound, an organosulfur compound, a nitrogen content compound, a tin system compound, organic peroxide, etc. can be used suitably. Still more specifically 2-benzothiazolyl sulfide, a benzothiazole, A thiazole, dimethyl acetylene die carboxylate, diethyl acetylene die carboxylate, BHT, butylhydroxyanisole, vitamin E, 2-(4-mol FOJI nil dithio) benzothiazole, 3-methyl-1-butene-3-ol, an acetylene nature unsaturation machine content ORGANO siloxane, Acetylene alcohol, 3-methyl-1-butyl-3-ol, diallyl fumarate, Diallyl maleate, diethyl fumarate, diethyl maleate, dimethyl maleate, 2-pentene nitril, 2, and 3-dichloropropene etc. is mentioned, and especially, in respect of coexistence of pot life/fast curability, although a thiazole and a benzothiazole are desirable, it is not necessarily limited to these.

[0024] Furthermore, to the hardenability constituent of this invention, other bulking agents, an antioxidant, an ultraviolet ray absorbent, a pigment, a surfactant, etc. can be added suitably if needed. As an example of this bulking agent, titanium oxide, a

zinc white, a barium sulfate, etc. can be mentioned, for example. The constituent of this invention offers the hardenability constituent by which adhesive improvement was carried out about the adhesive property made difficult from the former in the hardenability constituent using the saturated-hydrocarbon system polymer excellent in low moisture permeability, low hygroscopicity, low permeability, thermal resistance, weatherability, electric insulation, and oscillating absorptivity which can be hardened according to addition mold curing.

[0025] About low moisture permeability, the moisture permeation coefficient shows the outstanding property of 1×10^{-11} , cm/cm², and a sec-cmHg grade among many aforementioned properties. Moreover, about thermal resistance, 130 degrees C shows further an outstanding dielectric constant called about 2.44 in the outstanding property that melting is not seen on a front face on the severe conditions of 300 days about electric insulation.

[0026] The constituent of this invention can be used for broad applicable fields, such as closure material, such as electrical and electric equipment and electronic parts, various coating materials and the charge of gasketing, a sealing material, a molding material, a paint, and adhesives, from such a property. Below, based on an example, this invention is explained still in detail.

[0027]

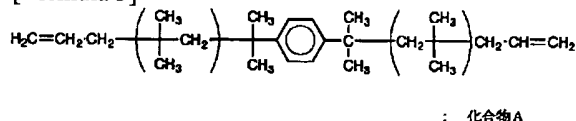
[Example] The molecular weight of the compound A shown in this example was measured by GPC and ¹H-NMR.

GPC analysis: -- system; -- Shodex by system made from Waters (pump 600E, differential refractometer 401), and column; Showa Denko K.K. Product made from 1 H-NMR: Varian which expresses K-804, mobile-phase; chloroform, number average molecular weight, etc. with polystyrene conversion Gemini-300, a measurement solvent; carbon-tetrachloride + pile acetone, and pressure-cooker processing -- Hirayama factory MODEL PC422R (serial No.901270620) was used.

[0028] The compound A which is the (A) component which shows structure to the following compounded by the method of a publication to example 1 JP,63-105005,A (an analysis value is shown in Table 1)

[0029]

[Formula 3]



[0030]

[Table 1]

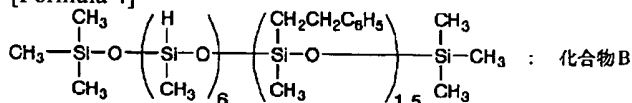
分析項目	測定方法	測定値
数平均分子量 (Mn)	G P C	5,400
重量平均分子量 (Mw)	G P C	7,200
分散度	G P C	1.34
末端官能基数	¹ H-NMR	1.99

注1) G P C測定値はポリスチレン換算値

注2) ¹H-NMR測定による末端官能基数は、開始剤残基に対する末端官能基の数を示す。

[0031] And the compound B which is the (B) component which shows structure below [0032]

[Formula 4]



[0033] Weighing capacity was carried out so that the ratio of the alkenyl radical weight of a ** (A) component and the Si-H radical weight in the (B) component might be set to 1:1.2, 1 weight section weighing capacity of IRUGA NOx 1010 (product made from Ciba-Geigy) was further carried out to the (A) component 100 weight section as an antioxidant, and this was kneaded 3 times with a roll. Then, it is the 4.4 weight sections and a (E) component to the (A) component 100 weight section about 3-glycidoxypopyltrimetoxysilane (the Nippon Unicar make, tradename:A-187) as a (D) component. 2.0 weight sections weighing capacity of the tetrapod ethoxy silane (the product made from Shin-etsu Chemistry, LS-2430) was carried out to the (A) component 100 weight section to the (A) component 100 weight section as the 0.2 weight section and a (F) component, and aluminum acetylacetonate was mixed. Furthermore, as a preservation stability improvement agent, to platinum, weighing capacity of the dimethylmalate was carried out so that platinum might become 5×10^{-4} Eq to the number of mols of the alkenyl radical weight of the (A) component about a screw (1, 3-divinyl -1, 1 and 3, 3-tetramethyl disiloxane) platinum complex compound catalyst (8.3×10^{-5} mmol/mu l, xylene solution), and it was uniformly mixed as a catalyst of the 30-mol equivalent and the (C) component. While investigating the hardenability of this constituent, a test piece is created, and it heated for 1 hour and was made to have applied on various base materials and to harden at 150 degrees C.

[0034] The test piece was produced for the addition of the condensation product (COL Coat, tradename:ethyl silicate 40) of a tetrapod ethoxy silane as the 2.3 weight sections to the (A) component 100 weight section like example 2 example 1 as a (F)

component.

The test piece was produced [titanium tetrapod butoxide] for the addition of the condensation product (COL Coat, tradename:ethyl silicate 40) of a tetrapod ethoxy silane as the 2.3 weight sections to the (A) component 100 weight section like example 3 example 1 to the (A) component 100 weight section as the 0.2 weight section and a (F) component as a (E) component.

[0035] The test piece was produced [titanium tetrapod butoxide] for the addition of the condensation product (COL Coat, tradename:ethyl silicate 40) of a tetrapod ethoxy silane as the 1.2 weight section to the (A) component 100 weight section like example 4 example 1 to the (A) component 100 weight section as the 0.2 weight section and a (F) component as a (E) component.

[0036] The test piece was produced like example of comparison 1 example 1, without adding the (F) component.

Like example of comparison 2 example 1, 2.0 weight sections addition of the titanium tetrapod butoxide was carried out to the (A) component 100 weight section as a (E) component, however the (F) component produced the test piece, without adding. Each combination was summarized into Table 2.

[0037]

[Table 2]

		実施例 1	実施例 2	実施例 3	実施例 4	比較例 1	比較例 2
(A) 成分 化合物 A	g	20	20	20	20	20	20
(B) 成分 化合物 B	g	1.36	1.36	1.36	1.36	1.36	1.36
(C) 成分 Plt ニモロチン錯体	μl	45	45	45	45	45	45
(D) 成分 3-オキシニトリロキシラン	g	0.89	0.89	0.89	0.89		0.89
(E) 成分 7-ニトリロキシラン	g	0.04	0.04	0.04		0.04	
チンチンチンチン	g				0.04		0.04
(F) 成分 チンチンチンチン	g	0.39					
スチレンチンチン	g		0.47	0.24	0.24		
貯蔵安定性改良剤 ジメチルチンチン	μl	14.1	14.1	14.1	14.1	14.1	14.1
酸化防止剤イタノックス 1010	g	0.2	0.2	0.2	0.2	0.2	0.2

[0038] They are an ordinary state and pressure-cooker (PCT) processing (121 degrees C, two atmospheric pressure.) about the test piece obtained as mentioned above about adhesive evaluation. The **** shear strength or the squares tape method of JISK6850 convention estimated the bond strength after 20-hour processing. Aluminum (thickness of 1.6mm) was used as a base material. Moreover, the squares tape method estimated glass (thickness of 2.8mm). The evaluation result about an adhesive property is shown in Table 3 and 4.

[0039]

[Table 3]

引張剪断強度 (kgf/cm2)

		実施例 1 kgf/cm2	実施例 2 kgf/cm2	実施例 3 kgf/cm2
アルミニウム	初期	9.6 A F	6.7 A F	7.3 A F
	PCT後	15.7 C F	20.6 C F	17.7 C F

		比較例 1 kgf/cm2
アルミニウム	初期	12.6 A F
	PCT後	15.7 A F

		実施例 4 kgf/cm2
アルミニウム	初期	4.5 A F
	PCT後	23.6 C F

		比較例 2 kgf/cm2
アルミニウム	初期	5.7 A F
	PCT後	16.2 A F

表中 C F は層間破壊、A F は界面破壊を表す

[0040] If the (F) component is not added from Table 3, it becomes interface destruction and it turns out that adhesive strength with a base material is small.

[0041]

[Table 4]

粘着試験

		実施例 1	実施例 2	実施例 3
ガラス	初期	25/25	25/25	25/25
	PCT後	25/25	25/25	25/25

		比較例 1
ガラス	初期	25/25
	PCT後	0/25

		実施例 4
ガラス	初期	25/25
	PCT後	17/25

		比較例 4
ガラス	初期	25/25
	PCT後	12/25

[0042] The numeric value in Table 4 puts the break of 25 2mm angles into a hardened material, and expresses the number which remained when it exfoliated on a tape. In 25/25, the number which did not exfoliate at all is expressed and it means that all 0/25 exfoliated. If the (F) component is not added from Table 4, it turns out that adhesive strength with a base material is small.

The ratio of the alkenyl radical weight of an example 5 (A) component and the Si-H radical weight in the (B) component should be set to 1:2.0. Weighing capacity is carried out and the (A) component 100 weight section is further received in a process oil (the Idemitsu Kosan make, tradename-S-32) as a plasticizer. 50 weight sections, furthermore -- as an antioxidant -- MARK AO-50 (product made from ADEKA AGASU Chemistry) -- the (A) component 100 weight section -- receiving -- 3 weight sections -- 100 weight sections weighing capacity of the fused silica (Made in Tatsumori) was further carried out to the (A) component 100 weight section, and this was kneaded 3 times with a roll. Then, it is the 4.3 weight sections and a (E) component to the (A) component 100 weight section about 3-glycidoxypyltrimetoxysilane (the Nippon Unicar make, tradename:A-187) as a (D) component. 1.9 weight section weighing capacity of the tetrapod ethoxy silane (the product made from Shin-etsu Chemistry, LS-2430) was carried out to the (A) component 100 weight section to the (A) component 100 weight section as the 0.2 weight section and a (F) component, and titanium tetrapod butoxide was mixed. Furthermore, as a preservation stability improvement agent, to platinum, weighing capacity of the dimethyl maleate was carried out so that platinum might become 5×10^{-4} Eq to the number of mols of the alkenyl radical weight of the (A) component about a screw (1, 3-divinyl -1, 1 and 3, 3-tetramethyl disiloxane) platinum complex compound catalyst (8.3×10^{-5} mmol/ μ l, xylene solution), and it was uniformly mixed as a catalyst of the 30-mol equivalent and the (C) component. While investigating the hardenability of this constituent, a test piece is created, and it heated for 1 hour and was made to have applied on various base materials and to harden at 150 degrees C.

[0043] The test piece was produced for the addition of a tetrapod ethoxy silane as the 1.0 weight section to the (A) component 100 weight section like example 6 example 5 as a (F) component.

The test piece was produced for the addition of the condensation product (COL Coat, tradename:ethyl silicate 40) of a tetrapod ethoxy silane as the 2.3 weight sections to the (A) component 100 weight section like example 7 example 5 as a (F) component.

[0044] The test piece was produced for the addition of the condensation product (COL Coat, tradename:ethyl silicate 40) of a tetrapod ethoxy silane as the 1.1 weight section to the (A) component 100 weight section like example 8 example 5 as a (F) component.

The test piece was produced like example of comparison 3 example 5, without adding the (F) component. The result of adhesive evaluation is shown in Table 5 about each combination in Table 6 and Table 7.

[0045]

[Table 5]

		実施例 5	実施例 6	実施例 7	実施例 8	比較例 3
(A) 成分 化合物 A	g	10	10	10	10	10
(B) 成分 化合物 B	g	0.93	0.93	0.93	0.93	0.93
(C) 成分 Plt ニオキチケル物	μl	22.1	22.1	22.1	22.1	22.1
(D) 成分 3-メチル-4-ヒドロキシベンゾイル	g	0.43	0.43	0.43	0.43	0.43
(E) 成分 テトラポッド	g	0.02	0.02	0.02	0.02	0.02
(F) 成分 シリカ	g	0.19	0.1			
硬化剤	g			0.23	0.11	
可塑剤 PS-32	g	5	5	5	5	5
フィラー 溶融シリカ	g	10	10	10	10	10
貯蔵安定性改良剤 ジメチルシリケート	μl	14.1	14.1	14.1	14.1	14.1
酸化防止剤 MARK AO-50	g	0.2	0.2	0.2	0.2	0.2

[0046]

[Table 6]

引張剪断強度 (kgf/cm²)

		実施例 5	実施例 6	実施例 7	実施例 8
		kgf/cm ²	kgf/cm ²	kgf/cm ²	kgf/cm ²
アルミニウム	初期	9.1 C F	8.9 C F	8.2 C F	9.4 C F
	PCT後	11.9 C F	12.0 C F	12.1 C F	12.4 C F

		比較例 3
		kgf/cm ²
アルミニウム	初期	9.7 C F
	PCT後	10.1 C F

表中 C F は層集破壊、A F は界面破壊を表す

[0047]

[Table 7]

各種目試験

		実施例 5	実施例 6	実施例 7	実施例 8
ガラス	初期	25/25	25/25	25/25	25/25
	PCT後	25/25	25/25	23/25	25/25

		比較例 3
ガラス	初期	25/25
	PCT後	22/25

[0048] The numeric value in Table 7 puts the break of 25 2mm angles into a hardened material, and expresses the number which remained when it exfoliated on a tape. In 25/25, the number which did not exfoliate at all is expressed and it means that all 0/25 exfoliated. If the (F) component is not added from Table 7, it turns out that adhesive strength with a base material is small. The following things became clear from the result of Tables 3, 4, 6, and 7. (E) The durable adhesive property of a hardened material and a durable bond strength improve by carrying out little addition of an organoaluminium compound or the organic titanium compound as a component, and carrying out little addition of tetrapod alkoxy silane or its condensation product as a (F) component. An effect is large to especially glass.

[0049] As mentioned above, the hardenability constituent by this invention has a scope wider than before, and its adhesive property improves.

[0050]

[Effect of the Invention] According to this invention, in the addition mold-curing system using the saturated-hydrocarbon system polymer, hardenability was not spoiled but it made it possible to raise an adhesive property so that clearly from an example. Especially the addition mold-curing object using the saturated-hydrocarbon system polymer is the rubber-like elasticity object excellent in various properties, such as low moisture permeability, low hygroscopicity, low permeability, thermal resistance, weatherability, electric insulation, and oscillating absorptivity, and can raise the reliability as closure material, such as the field by which an adhesive property is made indispensable, for example, the electrical and electric equipment, electronic parts, etc., various coating materials, a sealing material, a paint, adhesives, etc. by having an adhesive

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1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

CLAIMS

[Claim(s)]

[Claim 1] Hardenability constituent; containing the following component (A), (B), (C), (D), (E), and (F).

(A) The (curing agent C) hydrosilylation catalyst (D) silane-coupling-agent (E) organoaluminium compound which contains at least two hydrosilyl machines in the isobutylene system (polymer B) molecule which contains in a molecule the ARUKENIRU machine in which at least one hydrosilylation reaction is possible, organic titanium compound (F) multiple-valued alkoxy silane, and/or its condensation product. [Claim 2] (A) The hardenability constituent according to claim 1 whose polymer of a component is a polymer by which at least one ARUKENIRU machine is introduced into the end of a polymer chain.

[Claim 3] (A) The hardenability constituent according to claim 1 which is the polymer whose total amount of the repeat unit to which the polymer of a component originates in an isobutylene is 50 % of the weight or more.

[Claim 4] (A) The hardenability constituent according to claim 1 which is the polymer whose total amount of the repeat unit to which the polymer of a component originates in an isobutylene is 80 % of the weight or more.

[Claim 5] (B) The hardenability constituent according to claim 1 which is the polyorgano hydrogen polysiloxane to which the curing agent of a component contains at least two hydrosilyl machines in a molecule.

[Claim 6] (D) The hardenability constituent according to claim 1 which is a compound which has the alkoxy group which the silane coupling agent of a component combined with at least one functional group chosen from an epoxy group, an methacrylic machine, an acrylic machine, an isocyanate machine, an isocyanurate machine, a vinyl group, and a carbamate machine into a molecule, and silicon atom.

[Claim 7] (D) The hardenability constituent according to claim 1 which is a compound which has the alkoxy group which the silane coupling agent of a component combined with at least one functional group chosen from an epoxy group, an methacrylic machine, and an acrylic machine into a molecule, and silicon atom.

[Claim 8] (E) The hardenability constituent according to claim 1 whose organoaluminium compound of a component is an aluminum chelate compound.

[Claim 9] (E) The hardenability constituent according to claim 1 which is an organic titanium compound in which the organic titanium compound of a component has Ti-O-C combination.

[Claim 10] (F) The hardenability constituent according to claim 1 whose multiple-valued alkoxy silane of a component is tetrapod alkoxy silane and/or its condensation product or trialkoxy silane, and/or its condensation product.

[Translation done.]